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SCIENTIFIC BASIS FOR NUCLEAR WASTE MANAGEMENT XX

COMPARISON OF THE DISSOLUTION RATES OF URANIUM OXIDES IN AQUEOUS SOLUTIONS

The purpose of our work has been to measure the intrinsic dissolution rates of uranium oxides under a variety of well-controlled conditions that are relevant to a geologic repository and allow for modeling. The intermediate oxide phase, U_3O_8, is quite stable and known to be present in oxidized spent fuel. Dehydrated schoepite, $UO_3 \cdot H_2O$, has been shown to exist in drip tests on spent fuel.

Statistical experimental design was used to plan a set of 25 U_3O_8 and $UO_3 \cdot H_2O$ dissolution experiments under similar conditions. These experiments allow us to examine systematically the effects of temperature (25-75˚C), pH (8-10) and carbonate (2-200x10^{-4} molar) concentrations on dissolution of these oxides at 8 ppm dissolved oxygen in the leaching solutions, equivalent to atmospheric conditions.

Results indicate that $UO_3 \cdot H_2O$ has a much higher dissolution rate than U_3O_8. Dissolution of of both oxides show a high sensitivity to carbonate concentration and temperature. The activation energy for U_3O_8 dissolution was calculated as 6.1 kcal/mol by regression analysis of the data, using the classical chemical rate equation. This is equivalent to the previously reported activation energy for UO_2 at atmospheric conditions, within the error bounds of the measurements.

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